SEA-SALT OPTICAL PROPERTIES OVER THE REMOTE OCEANS: THEIR VERTICAL PROFILES AND VARIATIONS WITH WIND SPEED

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ABSTRACT

The size distribution and scattering coefficient of the marine aerosol was measured over the Southern Ocean during the First Aerosol Characterization Experiment (ACE 1) and over the tropical Pacific Ocean during the Second Pacific Exploratory Mission in the Tropics (PEMT-B). Thermal analysis of particles at near ambient temperature (40 °C), 150 °C, and 300 °C enabled us to estimate size distributions of the volatile (mostly sulfate and organic) and refractory (mostly sea-salt) modes as well as their contributions to optical depth and cloud condensation nuclei. When averaged spatially and temporally, sea-salt concentrations show dependence upon wind speed over the Southern Ocean up to an altitude of 1200 m. Their median contribution to column optical depth is 0.057, 0.033, and 0.021 at wind speed >10 m/s, 5 – 10 m/s, and ≤ 5 m/s, respectively, and the median over all wind speed categories is 0.041. However, on a case-by-case basis, the physical and optical properties show a large variance due to other meteorological conditions. The scales of this variability are also discussed along with examples of processes that contribute to them such as cloud processing and vertical wind speed gradient.
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1. INTRODUCTION

Aerosols are suspended particles in the atmosphere ranging from a few nanometers (nm) to tens of micrometers (µm) in diameter. The aerosols arise from natural sources (e.g., dust from deserts, soot from volcanoes, and sea salt from ocean surface) and anthropogenic sources (e.g., sulfate and elemental/organic carbon particles from biomass burning, fossil fuel combustion, and industrial processes). Once produced, aerosol concentrations are modified by various processes including long-range transport of continental air, precipitation, dry deposition, coagulation, and condensation. As a result, residence times of the particles often depend on their size and vary from a few hours to a few weeks, which leads to their inhomogeneous distributions in terms of time and space. This is in sharp contrast to the greenhouse gases such as carbon dioxide, methane, and nitrous oxide. These gases may remain in the atmosphere for a scale of 10 – 100 years, resulting in the more homogeneous distribution over the whole global atmosphere.

Aerosols can impact the Earth’s climate both directly and indirectly. In the so-called “direct effect”, the aerosols scatter and/or absorb the incoming solar (shortwave) radiation, thereby affecting the Earth’s radiation budget. Additionally, via the “indirect effect”, increases in the aerosol number may increase cloud droplet number, which can result in enhanced cloud reflectivity (albedo) [Twomey, 1974] as well as decreased precipitation efficiency and increased cloud lifetimes [Albrecht, 1989; Rosenfeld and Farbstein, 1992]. As a result, the aerosols are expected to result in a cooling of the Earth, except for highly absorbing aerosol species such as black
carbon [IPCC, 2001]. This effect is opposite to the warming effect due to the greenhouse gases [IPCC, 2001] but due to large gradients in aerosol concentrations regional effects may be much larger or smaller than those of greenhouse gases. Some absorbing aerosols can even change the atmospheric temperature structure and weaken the hydrological cycle [Ramanathan et al., 2001]. Yet, the existing estimates of these effects have large uncertainties.

Aerosols over the remote oceans are dominated by two major species, sea salt and sulfate, with a smaller and poorly characterized fraction of organic aerosols. The major components of sea salt are chloride, sodium, magnesium, sulfate, calcium, and potassium. The marine aerosols are an important contributor to the global aerosol budget and its optical effects, as the oceans cover approximately 70% of the Earth's surface. Considerable effort has been made to obtain their concentration, size, and chemical composition. In his early study Woodcock [1953] collected aerosol on small glass slides exposed from an aircraft. Reanalyzing his observation, Blanchard and Woodcock [1980] developed vertical sea-salt distributions for average wind speeds of 1, 3.5, 8, and 14 m/s. Their results are repeatedly used in later model studies (for example, [Collins et al., 2001; Gong et al., 1997; Porter and Clarke, 1997]). Blanchard et al. [1984] used the same technique on 8 different days and described the vertical structure of the sea-salt particles. Hoppel et al. [1994] sized particles that were sampled from an air ship in the marine boundary layer (MBL), and found that cloud processing affects the marine aerosol size distributions under nonprecipitating stratus. These and other previous attempts to measure sea-salt size distributions are summarized in the review by Fitzgerald [1991] and O'Dowd et al. [1997]. While sea-
salt particles are ubiquitous and dominant in mass over the oceans ([Quinn et al., 1998]), sulfate particles in the MBL also play an important role in modifying aerosol radiative effects. It has been argued that biogenic dimethylsulfide (DMS) emitted from the seawater to the remote marine atmosphere is a major source of marine sulfate and cloud condensation nuclei (CCN) [Charlson et al., 1987]. However, more recent measurements indicate that DMS oxidation results in heterogeneous deposition to existing aerosol surfaces in the MBL, while new particle formation takes place aloft [Clarke et al., 1998; Raes, 1995]. The optical properties of marine aerosol have also been studied using ground measurements (e.g., [Kaufman et al., 2001]) and models (e.g., [Takemura et al., 2000; Tegen et al., 1997; Uno et al., ]).

Some previous studies suggest that the vertical profiles of sea-salt are highly dependent on wind speed and other meteorological conditions related to sea-salt production, scavenging, atmospheric stability and convection [Blanchard et al., 1984; Exton et al., 1983; Fairall et al., 1983; Fitzgerald, 1991]. These studies are mostly theoretical, and only a limited number of actual observations of the sea-salt vertical profiles are available to support them. Although aircraft should be the best tool to observe the vertical structure, the previous aircraft observations mentioned above do not offer statistically robust information of the vertical profiles under a range of typical conditions.

Aerosol optical depth (AOD, defined in section 2.2) has a direct relevance to the study of radiation forcing. Kaufman et al. [2001] find from their automated ground measurement network that the baseline maritime AOD over the Pacific Ocean is 0.052 at 500 nm. Wilson and Forgan [2002] reported seasonal and yearly
variations of AOD over Tasmania at four wavelengths. It is important for field measurements to be capable of obtaining it with some accuracy. Surface measurements of AOD are column-averaged properties that reflect the size-resolved physical, chemical, and hygroscopic properties. However, attributing the contribution to AOD due to various constituents requires vertical evaluations of the related properties. Differential optical depth, as an integral of scattering coefficients over known altitudes (see the Beer-Lambert equation in section 2.2), provides a way to apportion AOD to various aerosol constituents. When integrated over the column flown in conjunction with contributions from higher altitudes these data can be compared to surface measurements.

Satellite observations are expected to improve our ability to study aerosols. Unlike flight campaigns and other in situ measurements, satellites can cover large scales in terms of both space and time. However, constraints based on observation are often required to obtain realistic aerosol properties, regardless whether they use spectral or angular scattering characteristics of aerosols. For example, the aerosol data retrieval algorithm for the moderate resolution imaging spectroradiometer (MODIS) [Kaufman et al., 1997] assumes 9 different aerosol models, each of which is associated with optical depth as a function of wavelength. The combination of the models that best fits the observed irradiance over multiple wavelengths is used to constrain the optical depth and other parameters. The models are based on sky radiance measurements of various aerosol species. Similar to model studies, the satellite studies to date have retrieved integral column properties of the aerosol but little is inferred about the vertical structure of the marine aerosol. Detailed
measurements of in situ vertical profiles under open ocean conditions are essential to support both model and satellite objectives.

During the First Aerosol Characterization Experiment (ACE 1) and the Second Pacific Exploratory Mission in the Tropics (PEMT-B), the aerosols in the clean MBL were sampled for about 110 hours in total. They provide statistically improved information of the aerosol vertical profiles and their physiochemical properties. This thesis will provide a quantitative assessment of the role of sea salt in modifying the optical properties of the MBL. Particular effort will be made to establish the vertical profiles of aerosol concentration and optical properties including their wind speed dependence. Other factors contributing to their variability will be discussed.
2. THEORETICAL BACKGROUND

2.1 Aerosol physical properties

The number size distribution of the aerosol is often expressed as $dN / d\log D_p$, or number of particles per cm$^3$ of air in the size range $\log D_p$ to $(\log D_p + d\log D_p)$ where $D_p$ refers to particle diameter in units of μm. The integral number concentration of particles is expressed as:

$$ N = \int_{-\infty}^\infty \frac{dN}{d\log D_p} d\log D_p \text{ (cm}^3) $$

When plotted versus $\log D_p$, the $dN / d\log D_p$ curve enables one to analyze a wide order of magnitude of particle sizes as well as to visualize the total number of particles which is the area below the curve (Figure 1).

Similarly, for the study of aerosol microphysics and chemistry, the surface area and volume distributions are defined as follows under the assumption of spherical particles.

$$ dA / d\log D_p = dN / d\log D_p * \pi * D_p^2 \text{ (μm}^2 \text{ cm}^{-3}) $$

$$ dV / d\log D_p = dN / d\log D_p * \pi / 6 * D_p^3 \text{ (μm}^3 \text{ cm}^{-3}) $$

The total area and volume of particles are

$$ A = \int_{-\infty}^\infty \frac{dA}{d\log D_p} d\log D_p = \int_{-\infty}^\infty \frac{dN}{d\log D_p} * \pi * D_p^2 * d\log D_p \text{ (μm}^2 \text{ cm}^{-3}) $$

$$ V = \int_{-\infty}^\infty \frac{dV}{d\log D_p} d\log D_p = \int_{-\infty}^\infty \frac{dN}{d\log D_p} * \pi / 6 * D_p^3 * d\log D_p \text{ (μm}^3 \text{ cm}^{-3}) $$

Since the particle area and volume are proportional to the square and the cube of the diameter, even a small number of large particles can contribute significantly to the
total area and volume (Figure 1). The mass is derived from the particle volume and density \( \rho \) (g \( \mu \)m\(^{-3} \)), which varies with size and composition.

\[
dM / d\log D_p = dV / d\log D_p \cdot \rho \text{ (g cm}^{-3}\text{)}
\]

Aerosol sizes are often identified by the geometric, optical, or aerodynamic diameter. For example, our Optical Particle Counter, which will be described later, sizes individual particles based on the light scattering, while the impactor cut-off size for a nephelometer is defined by the aerodynamic diameter that is related to how the particle behaves in an air flow. The aerodynamic diameter is assumed to nearly equal to the square root of the particle density (g/cm\(^3\)) times the geometric diameter [Peters et al., 1993]. Thus, 0.65 \( \mu \)m in geometric diameter for a particle of 2.2 g/cm\(^3\) density such as a dry sea-salt particle is equivalent to about 1 \( \mu \)m in aerodynamic diameter for spherical particles. In this thesis, it is assumed that the effective optical diameter is equal to the geometric diameter, since these were based upon optical instruments calibrated with spherical calibration aerosols and sea salt has a similar refractive index as the calibration aerosol and is nearly spherical.

It is often necessary to relate the diameters measured under dry conditions to those under ambient relative humidity (RH). Growth factor, \( G \), is defined as the ratio of diameter at a given RH to that at a reference RH. Based on Tang et al. [1997]'s study of water uptake by sea-salt particles, we get the growth factor shown in Figure 2a. There is a sudden rise in the growth factor near 45% RH where the sea-salt changes its phase between crystal and hydrate. The growth factor is about 2 at 80% RH with reference to 0% RH.
The relative scattering coefficient as a function of RH, \( f(RH) \), needs to be applied to the nephelometer scattering coefficient to obtain the scattering coefficient under ambient condition. From their observations on a beach, Clarke et al. [personal communication] found

\[
f(RH) = (1 - RH / 100)^{-0.39} \text{ for } RH > 45\%
\]

for marine aerosols (Figure 2b). This is just a rough approximate, as the actual \( f(RH) \) depends on size distribution. For \( RH \leq 45\% \), the scattering coefficient drops sharply due to the efflorescence effect of sea salt [Tang, 1997].

2.2 Aerosol optical properties

Attenuation of light by aerosols is described by the extinction coefficient defined as the fractional loss of light intensity per unit path length, and conventionally expressed as \( \beta_{ext} \) with a unit of \((\text{Mm})^{-1}\) (i.e., \(10^{-6} \text{ m}^{-1}\)). It is linked to macroscopic light intensity loss over a distance \( z \) by the Beer-Lambert law

\[
\frac{F}{F_0} = \exp(-\beta_{ext} z)
\]

where \( F_0 \) and \( F \) is the light incident flux at \( z = 0 \) and \( z \), respectively. The dimensionless product \( \tau = \beta_{ext} z \) is called the optical depth of the air mass. The extinction coefficient is the sum of the scattering coefficient and the absorption coefficient. For the sea-salt and sulfate particles described here, the scattering coefficient is approximately equal to the extinction coefficient, because their absorption is negligible.

The aerosol scattering coefficient can be computed from the number distribution using the Mie theory. This theory assumes spherical particles and that the
scattering coefficient in a given air mass is the sum of the scattering effect due to the individual particles. The scattering effect due to a spherical particle is expressed as the product of its cross sectional area and the scattering efficiency $Q_{\text{scat}}$. Thus,

$$b_{\text{scat}}(\lambda) = \frac{D_{p,\text{max}}^2}{4} Q_{\text{scat}} \frac{dN}{d \log D_p} d \log D_p \quad (\text{Mm}^{-1})$$

$Q_{\text{scat}}$ is a non-dimensional function of the refractive index and size of the particle as well as the wavelength.

Figure 3 illustrates the above equation and reveals the so-called Mie oscillation in the scattering efficiency (thin blue curve) of a dry sea-salt particle at a wavelength of 550 nm. This oscillation causes difficulty in the accurate calibration of optical instruments, since the calibration must assume a single valued dependency. When scattering efficiency is expressed per unit mass (thick blue curve in Figure 3), it is evident that particles smaller than 0.1 μm or larger than 10 μm are relatively unimportant in scattering at 550 nm unless there is a large mass fraction in these sizes. The scattering distribution (thick black curve in Figure 3) is given as a product of scattering efficiency (thin blue curve in Figure 3) and cross sectional area distribution (thin magenta curve in Figure 3). The area below the thick black curve represents the scattering coefficient.

The refractive index is a non-dimensional complex number dependent on the chemical species of the particles and the wavelength of light. For non-absorbing particles such as sulfate, sea salt, and water, the complex part of the index is zero. Since sulfate and sea-salt particles adsorb water with increasing relative humidity (RH), their refractive indices are functions of RH. Tang [1997] and Tang and Munkelwitz [1991] established the “partial molal refraction approach” to calculate
refractive indices for sulfate and sea salt under a given RH (Figure 2c). Because the sulfate present for most of these data was only partially neutralized by ammonium, the refractive index for an equal molar mixture of sulfuric acid, H$_2$SO$_4$, and ammonium bisulfate, (NH$_4$)$_2$SO$_4$, is used for the volatile component of the particles in this thesis. Its refractive index is believed to vary slowly down to very low RH, as the sulfate particle has a strong affinity for water molecules even under dry conditions. On the contrary, that for sea salt shows a sudden drop at about 45% RH. With decreasing RH, phase transition occurs near 45% RH and the sea-salt particles nucleate in solution and the water evaporates.

The light scattered by the aerosol depends on the wavelength. The Angstrom exponent, $\hat{a}$, describes the wavelength dependence of scattering coefficient and is given by

$$\hat{a} = -\frac{d \log \beta_{ext}}{d \log \lambda} \approx -\frac{\log(\beta_{ext1}/\beta_{ext2})}{\log(\lambda_1/\lambda_2)}$$

Since the scattering coefficient is also dependent on particle size (thick blue curve in Figure 3), the Angstrom exponent can provide a simple indication of the size distribution. A large Angstrom exponent is associated with a small average aerosol particle size. Figure 4 shows an example of the measured dry nephelometer scattering coefficients and Angstrom exponent over a vertical profile from December 9, 1995 during ACE 1. The relative increase in coarse marine aerosol near the surface results in lower values of the Angstrom exponents.
3. EXPERIMENT

ACE 1 and PEMT-B involved extensive level legs and vertical profiles in remote unpolluted oceanic regions for 60 and 50 hours, respectively. These data sets measured by multiple instruments enabled us to comprehensively describe marine aerosol properties under various meteorological conditions. Since the sampled air consisted mainly of sulfate and sea salt with minimum pollution, unambiguous discussions of the baseline maritime air can be made based on these data.

ACE 1 employed the National Center for Atmospheric Research C-130 aircraft to measure microphysical, chemical, and optical properties of tropospheric aerosols over the southwest Pacific Ocean (Figure 5), south of Australia in November and December 1995 [Bates et al., 1998]. Of the 18 flights excluding transit flights, about 60 hours was spent in the lowest 2000-m layer over the open ocean to quantify the characteristics of the natural marine aerosols in the minimally polluted atmosphere. These flights comprised about 70 column profiles to obtain the vertical structure of the aerosol properties. They also included many horizontal legs, each of which lasted typically for 30 minutes over a circle of 60 km in diameter. Six of these flights yielded two successful series of Lagrangian measurements. In each Lagrangian experiment, an air mass was tagged with smart tetroons [Businger et al., 1996] and sampled during sequential flights over ~30 hours.

The PEMT-B experiment employed the National Aeronautics and Space Administration P-3B aircraft to fly over the tropical Pacific Ocean (Figure 6) in
March and April 1999. Similarly to ACE 1, of the 15 flights excluding transit flights, about 50 hours was spent in the lowest 2000-m layer over the open ocean.

A description of the relevant data sets follows. All of the observed values (e.g., the number size distribution and the light scattering coefficient) reported in this thesis are those measured over the remote marine air, more specifically the lowest 2000-m layer over the Southwest Pacific Ocean and the tropical Pacific Ocean. All samples were measured under dry conditions. The size distribution and light scattering coefficient are corrected for ambient temperature and pressure so that differences evident from different altitudes reflect actual changes independent of volumetric changes. We have removed the data associated with pollution which is indicated by high ratio of number of heated (residual) particles to that of unheated (total) particles measured by a condensation nuclei counter. We also removed data associated with cloud, rain, and instrument problems.

The number size distribution was measured between effective optical diameters of $0.15 - 7.7 \, \mu m$ in dry conditions (RH ~ 20 %) with a laser optical particle counter (OPC) (Particle Measurement Systems LAS-X, Boulder, Colorado modified for 256 channel resolution). The OPC diameter was calibrated with polystyrene latex (PSL) spheres and glass beads. The aerosol was preheated to 40, 150, and 300 °C to infer aerosol composition from volatility [Clarke, 1991]. A sequential cycle through these temperatures took 2 – 3 minutes. Smaller sizes were also measured between a mobility diameter of $0.01$ to $0.25 \, \mu m$ in dry conditions with a Radial Differential Mobility Analyzer (RDMA). The particle counts in channels (0.15 to 0.25 \, \mu m) that overlap with the OPC are not included in this thesis.
The aerosol scattering coefficient was measured at three wavelengths (450, 550, 700 nm) under dry conditions (25% RH on average below 1000 m) every 15 seconds with an integrating nephelometer (Thermal Systems Incorporated, Model 3563) [Anderson et al., 1996]. This instrument is designed to obtain the scattering coefficient within a precision of 0.1 Mm\(^{-1}\). Occasionally, an impactor with an aerodynamic cutoff diameter of 1.0 \(\mu\text{m}\) was used for the purpose of measuring the scattering coefficient of submicron particles. To reduce mixed aerosol due to nephelometer residence time, the transitional data 20 sec before and 40 sec after each impactor switch were removed. An additional correction for truncation error due to angular nonideality [Anderson and Ogren, 1998] is considered later in this thesis.

When available, the chemically derived mass of sea-salt was estimated from ion chromatography analysis of sodium that was collected with filter samplers installed inside and outside of the aircraft [Huebert et al., 1998; Huebert et al., 1990]. The external sampler was used only once per flight during horizontal legs and required 30 minutes or longer to take one sample. The internal sampler was used more frequently during each flight but required similar integration times. The sea-salt mass is given as the sodium mass divided by 0.3061 which is the weight fraction of sodium in sea-salt particles [Seinfeld and Pandis, 1998]. The internal filter sampler undercounted particle mass larger than about 1 \(\mu\text{m}\) due to loss at the impactor and tubing plumbing. In order to correct for this factor, particles losses in the aircraft plumbing and the impactor were measured with an Aerodynamic Particle Sizer as a function of aerodynamic diameter during a post-field experiment [Huebert, personal communication] (Figure 7a), and the overall transmission efficiency was given as a
product of the plumbing and impactor transmission efficiencies. The external sampler did not suffer from this problem.

The column optical depth was measured near the aircraft several times during ACE 1 with a sunphotometer aboard the R/V Discoverer. This ship, operated by the National Oceanic and Atmospheric Administration, served as a mobile surface station and coordinated with the C-130 aircraft during the ACE 1 intensive campaign. The random uncertainty in the sunphotometer measurement is estimated to be ±15%, although a bias to higher values caused by invisible cirrus in the region may impact some measurements.

The wind speed was logged on the aircraft data system along with latitude, longitude, altitude, ambient temperature, ambient pressure, and ambient RH. Although near-surface wind speed is most important for sea-salt production, the wind speed around the aircraft at an altitude of 30 m or higher is used as replacement in this thesis. Wind speed at aircraft altitude is most frequently 10 m/s or higher near Tasmania over the Southern Ocean, due to strong westerly winds rarely obstructed by land. The wind speed over the tropical Pacific Ocean is often lower and rarely associated with frontal systems. The wind speed varies depending mainly on the distance to the subtropical high pressure system as well as the strength of local convection.
4. DATA CORRECTIONS, ADJUSTMENTS, AND INTERCOMPARISON

Most aircraft observations require corrections and adjustments before they provide data relevant to ambient conditions. The factors for which ACE 1 raw OPC size distributions had to be corrected and adjusted include particle loss in the tubing and the inlet (Community Aerosol Inlet, described later in this section) and the difference in temperature, pressure, and RH between the ambient air and the OPC. Of these factors, the temperature and pressure differences were corrected for by applying the ideal gas equation. Treatments for the rest of the factors are described in this section. The OPC optically effective size distributions and their integrals as a result of each correction are compared with observations from other instruments to confirm the consistency of the measurements, corrections, and adjustments. The nephelometer measurement is also corrected for the truncation error and adjusted from the instrument RH to ambient RH. The size distributions and scattering coefficients from PEMT-B are adjusted similarly. Although comparison of OPC optically effective size distributions with measured aerosol mass is one of the approaches discussed here, it is important to note that the focus of this thesis is aerosol optics. Unlike mass, the latter is less sensitive to contributions from aerosol larger than 5 μm or so.

4.1 Tubing loss correction for the ACE 1 OPC size distribution

The nephelometer sampled from the same inlet (CAI) as the OPC so that the only difference in the air paths from the ambient air to these two instruments was the
tubing between the CAI and the OPC. Hence the dry scattering coefficients calculated from the OPC size distribution can be compared to those measured by the nephelometer. The OPC size distribution at 40 °C was averaged over each leg, usually a 60 km diameter circle (Figure 5). This was used to calculate scattering coefficient at 450, 550, and 700 nm based on Mie theory. In this calculation, a refractive index of the calibration particles (PSL), 1.58, was used. While the actual refractive index for each component of the marine aerosol under dry condition is estimated to be smaller by 0.01 (Figure 2c), the calibration refractive index is preferred for the purpose of scattering closure. The OPC size distribution is calibrated to yield effective optical diameters, that is, the particle size if the particles had a refractive index of 1.58. Because scattering varies with particle refractive index, the geometric diameters of the marine aerosol are often different from their effective optical size measured by the OPC. However, by applying the refractive index of the calibration particle and integrating the OPC effective optical distribution, we obtain the total scattering of the aerosol seen by the OPC.

Comparison of the scattering coefficient inferred from the OPC size distribution and that measured by the nephelometer requires adjustment of measured scattering due to the truncation errors due to angular nonideality of the nephelometer [Anderson et al., 1996]. Typical adjustment involves multiplying the truncation correction factors by the measured scattering coefficients [Anderson and Ogren, 1998]. The factors are often expressed as functions of the Angstrom exponent, as the magnitude of the error depends on the particle size. While this correction method is shown to be good for a certain bimodal size distributions over the submicron diameter
range, it is not clear if it holds for the size distributions studied in this thesis over both the total and submicron diameter ranges. To avoid this uncertainty in comparisons, in this study, the integral scattering coefficient measured by the nephelometer, $\beta_{\text{scat,nephangle}}$, was calculated from the OPC effective optical size distribution as integrated scattering over $7^\circ - 170^\circ$ instead of $0^\circ - 180^\circ$ including an estimated correction for the non-lambertian illumination by the light source [Anderson et al., 1996; Quinn et al., 1995]. The difference between the total scattering efficiency and $\beta_{\text{scat,nephangle}}$ is large and variable for particles larger than about 1 $\mu$m (Figure 8). This is primarily because nephelometer truncation error mainly affects forward scattering. Large particles have the greatest size-dependent forward scattering relative to total scattering.

The scattering coefficient calculated from the OPC size distribution is highly correlated ($R^2 = 0.95$) with that measured by the nephelometer on a leg average basis at 450, 550, and 700 nm (Figure 9). Here the nephelometer measurement has been corrected for the truncation error using the OPC size distribution without tubing loss correction (small). The agreement in the Angstrom exponent calculated from the OPC size distribution and the nephelometer scattering coefficient at 450 and 700 nm is scattered around the 1:1 line (Figure 10). However, the OPC Angstrom exponent tends to be slightly smaller than the nephelometer value, suggesting somewhat fewer large particles in the OPC. A systematic difference is evident in the linear regression of the OPC scattering coefficient against the nephelometer scattering coefficient. The slope, which represents the total OPC/nephelometer scattering ratio, would be 1.0 under ideal circumstances. It is important to note that this ratio for the study is 0.85
at 550 nm, indicating that the OPC scattering coefficient is lower than the nephelometer direct measurement by 15%.

In the following discussion, possible causes of this difference are investigated and demonstrated to be mostly due to particle loss in the sample tubing and uncertainty in flow rate. The tubing transmission efficiency as a function of diameter is then estimated to correct the OPC size distribution.

The OPC detection limit could cause the low OPC/nephelometer ratio. The OPC detected light scattering by the particles of 0.15 – 7.7 μm in diameter, while the nephelometer measured scattering from all particles in its chamber. In order to estimate the scattering effect of the particles smaller than the OPC lower detection limit (0.15 μm), the scattering coefficient was calculated from the mean RDMA size distribution using the Mie calculation at a wavelength of 550 nm. At this time the refractive index of dry sulfate (1.41) was applied to all the particles regardless of their sizes. The scattering contribution by the particles smaller than 0.15 μm is less than 0.1% of the mean nephelometer scattering coefficient, regardless of the choice of refractive index value between 1.33 and 1.58. Hence, scattering by these small particles does not explain the 15% difference between the OPC and nephelometer total scattering coefficients.

Neither does the scattering by the particles much larger than the OPC upper limit (7.7 μm). Blomquist et al. [2001] confirmed that the particles larger than 9 μm in aerodynamic diameter were almost completely removed by the CAI before they reached the sample tubing and instruments inside the aircraft. This aerodynamic diameter is equivalent to a 7.7 μm or smaller geometric diameter, depending on
particle density. In other words, few particles larger than the OPC upper detection limit should have passed through the CAI to enter the nephelometer chamber.

Consequently, it appears likely that the low OPC/nephelometer scattering ratio is due in part to losses in sample tubing between the end of CAI to the OPC. This hypothesis is supported by the size dependency of the OPC/nephelometer scattering ratio. The OPC submicron scattering coefficient is calculated as the sum of the scattering due to particles smaller than 1 µm in aerodynamic diameter. Although uncertainties in particle density and the flow rate (about 30 L/min) make it difficult to explicitly confirm the target aerodynamic cut size (1 µm), for the nephelometer a 50% cut-off size of 0.72 µm in optical (geometric) diameter represents the target performance of the impactor (P. Quinn, PMEL, personal correspondence). The optical diameter of 0.72 µm is equivalent to 1 µm in aerodynamic diameter for a particle of 1.85 g/cm³ density, which is the average of those for dry sulfate (1.5 g/cm³) and sea-salt (2.2 g/cm³). The OPC submicron scattering coefficient is actually the product of the total OPC scattering distribution and the impactor transmission efficiency (Figure 7b). The impactor transmission efficiency used here is based on that for the Berner impactor studied by [Wang and John, 1988]. It was given against aerodynamic diameter so it had to be shifted to account for the difference between aerodynamic and optical diameter before being applied to this study.

The OPC submicron scattering coefficient is shown to be 93% of the nephelometer submicron scattering coefficients at 550 nm. This submicron OPC/nephelometer scattering ratio is higher than found for the total OPC/nephelometer scattering ratio (0.85). This implies that the OPC did not
undercount the submicron particles as much as the total particles. This is consistent with expectations for particle losses in tubes for these sizes. Large particles are removed more easily than small ones as they have large impaction and sedimentation losses. It is also possible that particles larger than the nominal impactor size cut reached into the nephelometer due to bouncing, although it is difficult to quantify this effect.

The tubing transmission efficiency (Figure 7b) was estimated as a function of particle diameter in such a way that scales the OPC scattering closer to the nephelometer scattering. The efficiency takes its lowest value (45%) at 7.7 μm but is almost 100% over the submicron diameter range. After the diameter was shifted to correspond to the optical diameter (the red curve in Figure 7b), the OPC size distribution was divided by this efficiency.

The submicron OPC/nephelometer ratio remained close to 1.0 (from 0.93 to 0.95) after the tubing loss correction. The uncertainty in the OPC flow rate (a difference measurement between two flow controllers each rated as ±3% flow uncertainty) and possibly other unknown factors should have resulted in the systematic 5% difference. A part of this difference may also be attributed to the uncertainty in the Mie calculation, especially due to the assumption that there is no organic compound. Therefore, the size distribution over the whole diameter range was further divided by 0.95 in order to raise transmission to the expected 100% for most submicron sizes and to better estimate transmission losses for the supermicrometer aerosol.
After these corrections for the tubing loss and flow rate uncertainty, the total OPC/nephelometer ratio at 550 nm increased from 0.85 to 0.95 (Figure 9b). This remaining 5% difference is within the uncertainty of the OPC calibration for large aerosol. The OPC/nephelometer regression slopes trend from 0.99 at 700 nm (Figure 9a) to 0.89 at 450 nm (Figure 9c) for reasons that are not clear although the nephelometer is least sensitive at 450 nm.

Another approach to confirm the OPC measurement and the correction is to compare the mass calculated from the corrected OPC size distribution at 300 °C with the dry sea-salt mass inferred from the internal sampler (green circle in Figure 11). To obtain the dry sea-salt mass estimated from the OPC size distributions, the average distribution during each of the internal sampler measurements was multiplied by the size-dependent transmission efficiency associated with the internal filter measurement (see Figure 7a and section 3) and the density of dry sea salt (2.2 g/cm³), before the distribution was integrated. They agree well both before and after the tubing loss correction ($R^2 = 0.85$ for both). The length of the error bars was calculated as ±25% based on roughly estimated uncertainties in the OPC calibration (5% for diameter and 10% for number), flow rate (5%), and sea-salt density (5%) (Table 1). Although the large uncertainty makes it difficult to assess the mass closure very precisely, the OPC size distributions adjusted by 5% to agree with the submicrometer nephelometer scattering remain consistent with mass closure.

Technically OPC distributions should have been corrected for the tubing loss before it is used to evaluate nephelometer truncation error. However, corrected OPC distributions are not available until the nephelometer scattering coefficient is
corrected for the truncation error and used to correct the OPC tubing loss. This circularity apparently allows more than one set of the corrections: If a larger OPC tubing loss correction is assumed, or if it is assumed that corrected size distributions have much more large particles, then a larger nephelometer truncation correction should have applied. By applying a larger tubing loss correction, reasonable closure between the nephelometer and OPC scattering coefficients would be achieved. However, in fact the truncation correction factors calculated from the corrected OPC distribution is larger than that calculated from the uncorrected OPC distribution only by 0.6%, which indicates that the assumption was reasonable. Also, the internal mass comparison after the correction resulted in a good agreement: the methodology resulted in the ratio of OPC-derived mass to the measured mass only slightly larger than 1.0. If a larger nephelometer truncation error was applied and a consequently larger OPC tubing loss correction, then the resulting OPC-derived internal mass after the tubing loss correction would have been much larger than the measured internal mass. Thus the methodology was the most consistent choice among the possible solutions for the circularity.

4.2 CAI loss correction for the ACE 1 OPC size distribution

Another major sampling loss aboard the aircraft is due to the Community Aerosol Inlet (CAI). This inlet system brings ambient air to the instruments aboard the aircraft along a 5-m-long tapered inlet intended to minimize sample losses. The CAI resulted in a sampling loss that is dependent on the particle size and the loss is 50% or more for aerosols larger than 3 μm in aerodynamic diameter [Blomquist et
More detailed intercomparison of the CAI and Low Turbulence Inlet (LTI) during a post-ACE 1 aircraft campaign, PELTI, near US Virgin Islands yielded the transmission efficiency under CAI RH against the aerodynamic diameters shown in Figure 12 [Huebert et al., personal communication]. The LTI enhances concentrations of large particles, and the ratio of the enhancement was estimated both theoretically and through empirical evaluations. The CAI transmission efficiency is the ratio of the size distribution measured behind CAI with an Aerodynamic Particle Sizer (APS) to the size distribution measured behind the LTI and corrected with the enhancement ratio. The ratio shows an unrealistic increase over diameters larger than 7 - 8 \mu m. This is due to an instrumental artifact where the recirculation of particles near the detection zone of the APS makes particles seem larger than the actual size and counted as particles \(>7 \mu m\). The curve over this range was replaced with an exponential decrease as rapid as the decrease evident over 4 - 6 \mu m, as is shown in Figure 12. It is also unknown whether the transmission efficiency determined during PELTI is applicable to the ACE 1 case. Dust particles were sampled in addition to sea-salt particles at 100 m or lower during PELTI, while most of the coarse mode particles were dominated by sea salt during ACE 1. Dust particles observed during PELTI may bounce on the inner wall of CAI more readily than sea-salt particles, which may result in higher transmission efficiency. In addition, uncertainty in the APS measurements and the particle loss in the plumbing as well as the LTI enhancement calculation on which the CAI efficiency estimate is based may make the uncertainty in the CAI efficiency estimate larger. Nonetheless, the PELTI intercomparison provides the best estimate of the CAI transmission efficiency at this
moment, and this work has to rely on this transmission efficiency to infer the ambient size distribution for this ACE 1 study.

The reasonableness of the CAI loss correction factor is evaluated by externally measured mass comparison in a similar manner to the internally measured mass comparison mentioned above. In order to produce size distributions expected at CAI RH, the OPC diameters had to be corrected for uptake of water at ambient conditions. The previously described OPC size distributions were multiplied by growth factors determined by the difference in CAI and instrument RH. The CAI RH was calculated based on the CAI temperature and the ambient RH, assuming that the water vapor was conserved from the ambient atmosphere, and is lower than the ambient RH typically by 5%. After this adjustment for the CAI RH, the largest ambient diameter calculated from the OPC diameter and the growth factor was 12.4 μm. The resulting measured size distribution was divided by the CAI transmission efficiency. The blue dots in Figure 11 compare the OPC-derived dry sea-salt mass after the correction for the CAI loss to the dry sea-salt mass obtained from the external sampler. The OPC-derived mass is from the average distribution at 300 °C in each of the horizontal legs when the external sampler was operational. Additional uncertainties in the CAI transmission efficiency (estimated at 28% for 2 μm particles and depending on particle size based on the variability in the PELTI measurement), RH (3%), and growth factor (10%) increased the total uncertainty from ±25% for the OPC-derived internal mass to ±61% for the OPC-derived external mass. Half of the data lie within the error bars, which seems to support the CAI loss correction. However, there are four data points where the OPC-derived mass is nearly half as
much as the external sampler mass. The difference may be attributed to particles larger than 10 μm that could not be reconstructed well from the OPC size distribution. As discussed later in section 9.1, a model comparison suggests that the difference between the modeled and OPC-derived ambient dry mass is about a factor of 2, depending on the actual size distribution. However, because this disagreement is dominated by larger particle mass that has a low scattering efficiency (Figure 3) the impact on estimated scattering extinction will be much smaller and probably less than 10% in most cases. This factor will remain uncertain until large particles can be measured more reliably.

4.3 Ambient RH adjustment for the ACE 1 OPC size distribution

The RH measured in the sample line near the OPC was lower than the ambient RH during the ACE 1, which made the particles smaller in the OPC than they were in the ambient condition due to water evaporation. The OPC diameters were, therefore, increased to ambient conditions using the growth factor discussed above (Figure 2a).

4.4 Summary of corrections to size distributions

Though numerous, the corrections and the adjustments are all founded on fundamental properties of the sampling system and are demonstrated to be reasonable by comparison of the resulting size distributions and their integrals with independent measurements. The impact of each of the corrections and modeling is shown as number and volume distributions averaged over the whole ACE 1 remote marine aerosol distributions in Figure 13a. These adjustments are primarily applied to
supermicrometer aerosol and do not significantly affect the total number of particles, which are dominated by small particles. On average, the total aerosol number remained almost same between the raw (34.7 cm$^{-3}$) and the tubing-loss- and flow-rate-corrected (36.8 cm$^{-3}$). With the CAI loss correction, this increased to 38.0 cm$^{-3}$ and this number remained the same after the RH adjustment.

Despite the small change in total number, these corrections strongly affected the scattering coefficient. The derived scattering coefficient increased from 6.8 Mm$^{-1}$ to 7.6 Mm$^{-1}$ (1.1 fold) with the tubing loss and flow rate adjustment, to 11.5 Mm$^{-1}$ (1.7 fold) with the CAI loss correction, and to 35.2 Mm$^{-1}$ (5.2 fold) with conversion to ambient RH (Table 2). The uncertainty in the ambient OPC scattering coefficient is ±44%.

The size distributions obtained during heated scans were corrected and modeled in the same manner as the unheated scans. This operation is based on an external mixing assumption between the sulfate and sea-salt modes. If the refractory sea-salt particles were originally coated by volatile species (internally mixed), then the CAI loss efficiency should have been applied to the original (larger) diameter. The validity of this assumption remains uncertain until the mixing status is determined. However, in most cases the total particle numbers in sequential scans at higher temperature were lower than those at low temperature. This is consistent with the assumption that volatile (e.g., sulfate) particles are counted only in unheated scans, while refractory (e.g., sea-salt) particles are counted in all scans.
4.5 Truncation correction and RH adjustment for the ACE 1 nephelometer scattering coefficient

Our assessment of ambient scattering is based on the size-resolved measurements and adjustments discussed above to the 3-minute OPC data. However, the measured nephelometer dry scattering coefficient is valuable because it was measured at a higher temporal resolution. Since the nephelometer provides only an integral property over all sizes, it is impossible to apply the CAI loss transmission efficiency and RH growth factors to correct dry nephelometer scattering values to ambient. Therefore, the relative increase in the derived OPC scattering coefficient due to the CAI loss correction (a factor of 1.55 on average) was used to scale the nephelometer scattering coefficient below 2km. Similarly, the magnitude of the nephelometer truncation error was estimated using the ratio of the total OPC scattering to the scattering seen by the nephelometer (1.17). The nephelometer total scattering at 550 nm was increased by these factors to yield approximate dry scattering coefficients corrected for the truncation error and the CAI loss. They were further adjusted to the ambient RH using $f(RH)$ (Figure 2b). The dry RH was assumed to be the average value of 25%.

The resulting ambient nephelometer scattering coefficient is compared with the ambient OPC unheated scattering coefficient (Figure 14). The mean nephelometer scattering coefficient during 1 minute before and after each OPC measurement is plotted. While most of the dots are located near the ideal $y=x$ line, some dots are far from this relationship. Most of the outliers occur under dry conditions at high altitudes where the $f(RH)$ value for marine air mass may not
appropriate. Figure 15 shows an example of the consistency generally observed between the vertical ambient scattering coefficients derived from the nephelometer and the OPC.

4.6 Correction and adjustment for the PEMT-B OPC size distribution

The tubing loss correction and modeled growth to ambient RH were executed for the PEMT-B OPC size distribution in the same manner. A solid diffuser, instead of CAI, was used as an inlet during PEMT-B, and its efficiency given by Huebert et al. [personal communication] (Figure 12) was used to correct these data. This correction is smaller than the CAI loss correction for the ACE 1 size distribution. The average number and volume distributions after each of the corrections are shown in Figure 13b. The sudden decrease in the raw size distribution over the diameter range larger than 4 μm is due to a different upper limit on the OPC electronics for PEMT-B, and correspondingly the large diameter end of the corrected distribution (red curve) is not reliable. Also, the shapes of the distributions over supermicron diameter range are different between ACE 1 and PEMT-B. Since the shapes over this diameter range associated with Mie oscillation is sensitive to the calibration curves, even a subtle difference in ACE 1 and PEMT-B calibration curves result in considerable difference in the distribution. These factors result in large loss in mass in PEMT-B as well as an inability to identify the peak diameters in the actual distributions, but they are still useful in assessing the wind speed dependency, relative importance of submicron/supermicron particles, and geographical features.
5. AVERAGE PHYSICAL AND OPTICAL PROPERTIES AND THEIR
DEPENDENCE ON WIND SPEED

Description of the remote marine aerosol begins with its average properties
categorized by altitude and wind speed. Since the emphasis here is on the optical
properties, the description of the size distribution is used not to quantify the total
aerosol mass loading but to assist in understanding scattering due to each component
and size of the particles. The magnitude of underestimate of scattering due to large
particles is discussed in section 9.1.

The reconstructed ambient size distributions described earlier are categorized
into three wind speed (WS) categories: WS > 10 m/s (for most of the cases below 20
m/s, hereafter high wind), WS = 5 – 10 m/s (medium wind) and WS ≤ 5 m/s (low
wind). The near-surface wind speed is used to categorize the ACE 1 size distribution
and scattering coefficient. The near-surface wind speed is defined as the average
wind speed measured from the aircraft while it was below 200 m. Individual size
distributions collected at times greater than 2 hours before or after a near surface wind
measurement do not appear in this section. On the other hand, the local wind speed
around the aircraft at a given time is used to categorize the PEMT-B size distribution
and scattering coefficient. This is because the near-surface measurements were not
done with sufficient frequency to provide adequate information of the near-surface
wind speed.
5.1 Near-surface size distribution

For each wind speed category, the ACE 1 ambient OPC size distributions were averaged at altitudes of 200 m or lower for all flights near Tasmania (Figure 16a-d). The unheated RDMA distribution is also shown. The dip near 2–3 μm is an instrumental artifact due to Mie scattering oscillation where more than one size corresponds to a single scattering coefficient. In all the categories, the 40 °C and 300 °C curves almost overlap for diameters larger than 1 μm except for the curves in the high wind speed case where poor counting statistics resulted in a different distribution at the larger sizes. Variations in concentrations evident above 2–3 μm are expected due to low counting statistics at these sizes and a small difference in the flow path for each thermal range. This is also true for most of the individual cases, some of which are shown later in this thesis. Most of the large particles are refractory, as expected for sea salt. In contrast, for diameters 0.15 – 0.4 μm, large differences between the unheated and 300 °C curves are evident. Most of the particles in this size range are volatile, as expected for sulfate and some organic aerosols. However, there are a considerable number of refractory particles in this diameter range as well.

It is also evident that when averaged spatially and temporally during ACE 1 the number of the supermicron sea-salt particles is a strong function of wind speed. Figure 17 compares the mean number and scattering distributions for refractory sea-salt aerosol remaining at 300 °C in each of the wind speed categories below 200 m. The scattering of the refractory particles under high wind speeds is 3.5 times as large as that under low wind speeds. That under medium wind speeds is 2.4 times as large as that under low wind speeds. This average behavior demonstrates that large-scale
regional characteristics are consistent with the widely accepted view of the sea-salt particle production mechanism [Blanchard and Woodcock, 1980; Twomey, 1977]: Wind induces wave breaking and enhances bubble bursting, which produces new sea-salt particles in the atmosphere at the ocean surface. Note that wind speed near the ocean surface prior to the moment of the measuring is an important factor that determines the aerosol loading as discussed later, and that these average dependency of size distribution on local wind speed discussed here is likely to result from the fact that the near surface wind speed is most likely to remain constant.

The application of thermal analysis to the OPC data reveals that even over the submicron diameter range, sea salt (300 °C) is a significant component. In the diameter range from 0.15 to 0.4 μm, 11% of the particle number is from sea salt (Table 3) and this ratio increases to 20% for the diameter range from 0.15 to 1 μm. The number of submicron sea-salt particles is also dependent on the wind speed. More than 3 times as many submicron sea-salt particles larger than 0.15 μm exist under high wind speeds (15.3 cm⁻³) as under low wind speeds (4.5 cm⁻³). The abundance of sea salt relative to all particles also varies significantly: Refractory sea salt represents 28%, 19%, and 11% of total particle numbers over the diameter range of 0.15 – 1 μm for WS > 10 m/s, 5 – 10 m/s, and ≤ 5 m/s respectively (Table 20).

This major dependence of the number of submicron sea-salt particles on wind speed is in sharp contrast with that of volatile particles. Interestingly, the concentration of particles below 0.15 μm actually decreases with increasing wind speed by about 50%, indicative of a different source and argued to result from entrainment from above the MBL [Clarke et al., 1998].
In PEMT-B, sea-salt volumes (Figure 16e – h) are smaller and less variable between different wind speed categories than those in ACE 1. The relative volume of refractory sea-salt aerosol under high and medium wind categories are factors of 1.8 and 1.2 greater than low wind categories (Table 4). Unlike the average properties for ACE 1, the number of refractory particles and its ratio to the total particle number over the diameter range smaller than 0.4 μm is smaller under high wind speeds: The refractory particle number is 9.4 cm$^{-3}$ (6% of the total number) under high wind speed and 13.1 cm$^{-3}$ (13% of the total number) under low wind speed. Also, there are many more volatile submicron particles than ACE 1. Possible causes of these differences between ACE 1 and PEMT-B are discussed later.

These characteristics of the marine aerosol near the surface are reflected in the scattering distributions. Figure 18 shows the overall scattering distribution below 200 m and those categorized by wind speed. While volatile particles are dominant in the marine aerosol number, the light scattering is largely affected by the abundance of sea salt in the ACE 1 distributions. However, in PEMT-B, scattering by submicron particles is considerable, especially for medium and low wind categories. In fact, scattering due to particles between 0.15 and 1 μm is 22.8, 40.5, and 49.6% of total scattering in the high, medium, and low wind categories, respectively.

5.2 Vertical profiles of sea salt size distribution

Profiles of refractory sea-salt size distributions were made both by continuous descents and stepped layers at selected altitudes. Figure 19(a-d) and 20(a-d) show the same size distribution plots as Figure 16(a-d) for the altitudes ranging 200 – 600 m.
and 600 – 1000 m for ACE 1. When categorized by wind speed, supermicron sea-salt particles show almost the same rate of decrease with altitude. For example, the sea-salt number at 600 – 1000 m is about two thirds of that near the surface under all wind speed categories. The number of small sulfate particles remains almost constant among these three altitude bins for all wind speed categories.

In PEMT-B (Figure 19(e-h) and 20(e-h)), the decrease in the volume of supermicron sea-salt particles with altitude is less evident than that is in ACE 1. For example, the supermicron sea-salt volume under medium wind speed remains almost the same for 0 – 200 m (Figure 16g), 200 – 600 m (Figure 19g) and 600 – 1000 m (Figure 20g).

5.3 Vertical scattering profile and column optical depth

The observations above are based on the OPC size distributions and thermal analysis. The ambient scattering coefficient was reconstructed from the unheated size distribution for the marine aerosol during ACE 1 as described earlier and is collectively plotted against the altitude (Figure 21). The scattering coefficient is highest near the surface where sea-salt particles are abundant and it diminishes with altitude. The median value in each 200-m altitude bin (large dot) shows a gradual decrease in scattering up to about 1400 m, and is almost constant at higher altitude. It is inferred from this that most of the MBLs are less than 1400-m thick.

Figure 22 shows the scattering coefficient of the refractory particles measured by the OPC at 300 °C. There are too few data under low wind speed to calculate the
median optical depth. For the other wind speed categories, the optical depth by the refractory particles, mostly sea salt, is about 75% of the optical depth by all particles.

The size dependent corrections made for the OPC data can also be used to estimate correction for the nephelometer data as discussed above. This provides a means for correcting the nephelometer to ambient conditions in order to take advantage of its high temporal resolution. The nephelometer ambient scattering coefficient during ACE 1 is plotted against the altitude in Figure 23. The large green dots are again the median, not the average, of the scattering coefficients in the 200-m altitude bins. Note that on a case-by-case basis this nephelometer scattering coefficient is not as reliably corrected as the OPC scattering coefficient. Even so, the overall characteristics revealed by both of the above approaches are quite consistent.

The nephelometer ambient scattering coefficients below 1000 m are greatest under high wind speeds, with a median scattering coefficient in the lowest 200-m altitude bin of 72 Mm\(^{-1}\). This is much larger than those in medium (40 Mm\(^{-1}\)) and low wind (19 Mm\(^{-1}\)) categories. The median values vs. altitude also show a thicker (~500 m) MBL under high wind than under medium wind speeds (~300 m). There is no similar well-defined lower layer under low wind speeds. Integrating the scattering coefficient over altitude yields the optical depth. The median differential optical depth for the lowest 2000-m layer is 0.041. When classified by wind speed, the value is 0.057 for high wind, 0.033 for medium wind and 0.021 for low wind speeds. These values are higher than the optical depth measured at Cape Grim with a sunphotometer. Based on the observations by Wilson and Forgan [2002], the optical depth averaged over November and December 1986-1999, except for the period affected by the
Mount Pinatubo eruption, is $0.043 \pm 0.006$. Note that this value is a result of interpolation over the measurements at 500 nm and 778 nm, and that this average value over 10 years does not necessarily reflect the optical depth in 1995 when ACE 1 took place. There is a 37% difference between this literature value and the ACE 1 mean optical depth (0.059) that is a sum of the contribution from the lowest 2000-m layer (0.041) and that from the layer over 2000 - 10000 m (0.0177).

Thus the vertical scattering profiles during ACE 1 confirmed that high wind speed is associated with high scattering coefficient. However, even in a wind speed category, the scattering coefficient at a given altitude in the lowest 1000 m can have a wide range of values. For example, the scattering coefficient at 500 m under high wind can be 10 or 150 Mm$^{-1}$. Thus, wind speed alone is not an adequate predictor for the sea-salt contribution to scattering at any given altitude or point in time. The sources of this variance are discussed in the following sections.
6. OPTICAL DEPTH COMPARISON

In order to link the \textit{in situ} data sets to satellite observations it is necessary to integrate the optical depth over the vertical column. In this section, the optical depth derived from the OPC measurement is compared to the ship- and ground-based measurements to discuss consistency and error sources of the OPC-derived optical depth.

The OPC-derived ambient optical depth was compared to the column optical depth measured from R/V \textit{Discoverer} during ACE 1 Flight 17, 22, and 24 (Figure 5). The scattering coefficient was calculated from the ambient size distribution using the refractive index of sea salt given as a function of the ambient RH (Figure 2c). Integrating the scattering coefficient over vertical columns from the surface to 2000 m provides the differential optical depth. Although the lowest 2000 m usually has the highest scattering coefficients, the column optical depth requires the addition of contributions from altitudes above 2000 m. The first contribution is 0.0089 from altitudes of 2000 – 6000 m. This is assumed to be as large as the median of all measured ambient scattering coefficients integrated over 2000 – 6000 m. The second contribution is 0.0051 from 6000 – 10000 m. This number is obtained from the contribution from the 2000 – 6000 m layer based on the assumption that the scattering coefficient between 2000 – 10000 m maintains a constant mixing ratio and scales with the atmospheric pressure. And, the third number is 0.008 as contribution from the stratosphere, which is obtained from the stratospheric measurements in December 1995 by SAGE II at 550 nm (M. Sato, SAGE II Stratospheric Aerosol Optical
Thickness, 2002, available at http://www.giss.nasa.gov/data/strataer/, hereafter [Sato, 2002]). These are the closest data for contributions above 2000 m that were available but may not always be representative of where we flew.

Figure 24 shows the optical depth measured in the columns for three profiles flown near the R/V Discoverer. As Porter et al., [2001] pointed out, optical depths measured with a hand-held sunphotometer are frequently biased toward the larger side due to the difficulty to direct the instrument right to the direction of the sun. I took only data sets with 3 or more measurements within a few hours and removed extraordinary large optical depth relative to the other optical depths in the same set, before the average in Figure 24 were calculated. Although there are several more sets of the ship sunphotometer data, they have been removed due to insufficient number of measurements within a period up to a few hours. Note that in this figure both the OPC-derived optical depth and that measured from the ship are at a wavelength of 500 nm. Also shown in this figure is the Cape Grim optical depth at 500 nm averaged over December in 1986 – 1999 excluding 1991. The optical depth at 550 nm contributed from the stratosphere ([Sato, 2002]) averaged over the same period of time, 0.0014, has been added to the optical depth to top of troposphere at 500 nm (0.0037) under the assumption that this slight difference in wavelengths does not significantly affect the estimate of the stratospheric contribution. Two out of the three resulting OPC-derived column optical depths lie near the ideal 1:1 line and suggest that this analysis is consistent with observations. The other data point indicates larger sunphotometer optical depth. This may be due to the measurement difficulty mentioned above. However, high invisible cirrus was not uncommon
during ACE 1 and may also bias column AOD to higher values than due to aerosol alone.

In addition to the measurements aboard the ship that was located close to the aircraft during the ACE 1 experiment, a ground measurement over a longer time period is also available to be compared with the \textit{in situ} data. Optical depths measured at Cape Grim, Tasmania at about 90 m high for more than a decade is provided by Wilson and Forgan [2002]. For the period 1986 – 1999 except for the period affected by Mount Pinatubo, the optical depth averaged over November and December are 0.044, which was interpolated from the optical depths measured at 500 and 778 nm. Note that the average contribution from the stratospheric aerosol for this time period, 0.015, has been removed from this literature value, so the average optical depth integrated up to top of the atmosphere is 0.059. The ACE 1 nephelometer ambient scattering coefficients shown in Figure 23 were integrated over altitude up to 2000 m for each wind speed class, and plotted against wind speed (dots in Figure 25). The blue variance bars indicate the calculated variance within each wind class. To be compared with the Cape Grim measurements, these differential optical depths have to be adjusted for the possible scattering deficit due to large particles not being measured (see Figure 31 and discussion in section 9.1) and for the contribution from particles between 2000 m and the stratosphere. This possible deficit of 20% due to coarse particles scatter was added to the median optical depth values (indicated as dark blue bars in Figure 25). For the contribution from the altitudes 2000 – 10000 m and stratosphere, 0.014 and 0.008 were added for the same reasons discussed earlier in this section. The resulting column aerosol optical depth up to top of atmosphere is
0.047 for low wind speed, 0.062 for medium, and 0.090 for high (bars in Figure 25). The overall median optical depth turned out to be 0.063, which is only slightly larger than the Cape Grim average value of 0.059. This difference is attributed to the meridional difference in scattering coefficient in MBL. Figure 26 shows the nephelometer ambient scattering coefficient at 200 m or lower collectively plotted against the latitude. The flight measurements over the upwind region of Tasmania between 40 and 44 °S found the wind speed and scattering coefficient distinctively lower than they are over the regions to the south. This could result in a lower scattering coefficient and AOD at Cape Grim compared to the ocean to the south, which is qualitatively consistent with the results above.
7. LAGRANGIAN STUDIES OF AEROSOL EVOLUTION

The collected examples of ACE 1 measurements of the size distribution and scattering coefficient shown in Figures 16, 19, and 20 exhibit a large variance in each wind speed category. Hence, it is difficult to predict the scattering coefficient and vertical profile based solely on the wind speed at a given time. This variability is due to variations in production, removal, or advection of aerosol into the air mass for a given wind speed class. If one samples the same air mass repeatedly over time (Lagrangian) it is possible to remove uncertainties associated with advection and study the production and removal processes. Each of two successful Lagrangian flight series during ACE 1 observed an air mass tagged with smart tetroons [Businger et al., 1996] over 30 hours each. Unlike most of the other measurements, the continuous history of the meteorological conditions of those two particular air masses were recorded.

7.1 Constantly high wind case (Lagrangian A)

During ACE 1 flights 18 – 20 (Lagrangian A), the aircraft followed an air mass for 30 hours, flying in stacked circles (Figure 27a) advected with the wind. The northern part of the circles was under or in thick clouds that existed at 800 – 1000 m, while the southern part was relatively clear. For the entire period, the wind below 2000 m was between 10 – 20 m/s and fell into our high wind speed category.

The scattering distributions averaged over 200-m altitude bins are shown in Figure 28. For the first two stacks of legs the light scattering gradually increased at
altitudes lower than 1000 m, but the scattering decreased at 800 – 1000 m for the third stack (Figure 28c), and at 600 – 1000 m for the fourth stack (Figure 28d). This is because the air mass encountered the clouds at these altitudes and the particles were coagulated to result in different size distribution. In the meantime, the light scattering kept rising at altitudes lower than 600 m. The effect of the cloud processing is much less at the low altitudes. Another possibility is that after the cloud processing, the aerosol loading in the lower layer recovered most quickly. After this event, the light scattering again increased from 4th to 5th stacks between 10:00 and 17:30 at all the altitudes. Thus removal processes contribute to the variance in a given wind speed category. This process can also affect the aerosol differently at different altitudes such that column average properties can differ from near surface behavior.

7.2 Medium to high wind case (Lagrangian B)

ACE 1 Lagrangian B (flights 24 – 26) included a period of steady increase in wind speed associated with an increase in light scattering due to sea salt production. The air mass experienced near-surface wind speeds of about 6 m/s at around 12:00 that increased to a wind speed of about 10 m/s at around 15:00 (Figure 29). Despite this wind speed increase, the vertical scattering coefficient remained almost constant at 30 – 35 Mm⁻¹ up to 600 m over 0 – 2000 m. It was not until the near-surface wind speed increased again to 12 m/s between 15:00 and 21:00 that the near-surface scattering coefficient increased to about 70 Mm⁻¹. After 21:00 the wind speed stays nearly constant until 00:00 but the near-surface scattering coefficient continues to increase gradually. The differential optical depth as an integral of the scattering
coefficient over 0 – 2000 m showed the similar behavior: It remained nearly constant at 0.03 during until 15:00 even with the wind speed increase, and increased to 0.05 between 15:00 and 21:00. Thus the wind speeds below 200 m alone cannot explain this behavior.

The key to explain this aerosol evolution is the vertical wind speed gradient. Before 15:00 the lower wind speed and absence of a gradient is consistent with low sea salt production and turbulent mixing. This resulted in little transport of the sea salt from the ocean surface to above. A strong wind speed gradient is evident between 21:00 and 00:00. The enhanced vertical mixing transports the sea salt to above more rapidly, which is why at this time the scattering coefficient at 200 m is as high as 70 Mm$^{-1}$ or higher. The scattering coefficient at 600 m also started to increase after the wind speed gradient at this altitude became strong between 21:00 and 00:00. This increased vertical mixing would eventually result in a constant scattering coefficient over altitude up to 600 m after another several hours if the turbulent mixing remains the same. Thus, the gradient in the vertical scattering coefficient is controlled both by the near-surface wind speed and the gradient in the wind speed that drives the mixing process. In addition, continued high winds at the sea surface will increase the aerosol generation as the wave field responds slowly to increased wind speeds. The sea-salt evolution depends on how long the wind blows over the sea surface, and therefore, a time lag between a wind speed increase and increase in the sea-salt concentration is expected.
8. UNCERTAINTY ANALYSIS FOR SPATIAL SAMPLING

It is clear from the discussions above that significant variation in aerosol properties is present over small scales (~10 km). The spatial scale of aerosol properties is of interest in obtaining representative aircraft data. Patchiness in the aerosol distribution will influence the strategy for sampling a region such as the length and type of sample legs and the appropriate scales needed to compare to other platforms or satellite products. In comparing satellite observation with in situ measurements, it is important to know the spatial scale the measured aerosol optical properties should be averaged over. This is particularly true for obtaining representative profiles designed to address column properties, such as those needed for column closure.

Measurement sequences comprised of one descending profile and a few horizontal legs were frequently flown during ACE 1 over a period of about 3 hours or less. Twenty-three of these sets enabled analysis of spatial variability in the ACE 1 MBL. Figure 30a shows one example of the aircraft path. A horizontal leg typically tracked a 60-km diameter circle in 30 minutes, and the set of stacked legs (circles) were flown within 180 minutes from the initial descent.

The mean of the nephelometer ambient scattering coefficient during each leg was assumed to be representative of the region at the respective altitude. The difference between this mean value and the scattering coefficient measured at the same altitude ± 50 m during the initial descent is a measure of the deviation of a single profile value from the value most representative of the region. Figure 30b
shows the frequency distribution of this difference. The difference between the measurements made at altitudes during the initial descent corresponding to those of the horizontal legs is 15.5 $\text{Mm}^{-1}$ or less for most of the cases. This variance should include some variability due to time difference between each set of scattering measurements. Note that this variance is much larger than the precision of the nephelometer, 0.1 $\text{Mm}^{-1}$. This suggests that in comparing the spatially averaged scattering coefficient, such as a satellite measurement, to the aircraft *in situ* measurement at one point, a difference of 15.5 $\text{Mm}^{-1}$, which is equivalent to 35% of the median scattering coefficient below 1000 m, is to be expected over this region in the Southern Ocean as seen in the natural variability present during ACE 1.
9. COMPARISON WITH MODELS

9.1 Porter and Clarke number distribution model

The size distributions presented so far are based on the OPC measurements during two aircraft campaigns, and it of interest to see how these data compare with the previous measurements in other parts of the world. Also, the different techniques used in previous studies offer a possible estimate of the coarse particle deficit discussed in this thesis. Porter and Clarke [1997] suggested a sea-salt coarse mode size distribution based on their ship measurements (~20-m altitude) combined with those performed by Woodcock [1953] (~500 to 800-m altitude). They chose OPC dry aerosol distributions in the MBL that roughly correspond to the coarse mode size distributions measured by Woodcock over 0.8 – 1 μm. The ACE 1 distributions at 300°C at 500 - 800 m under wind speeds that fall into each of the categories in the Porter and Clarke model are compared. The Porter and Clarke model was developed based on dry particles, so in order to compare their results under ambient conditions the diameters for the Porter and Clarke model distributions have been shifted by a growth factor of 2 that accounts for water uptake due to an RH shift from 0% to 80%.

The refractory ambient ACE 1 distributions for wind speeds of 5.5 – 7.9 m/s and 13.9 – 17.1 m/s fall close to the Porter and Clarke distributions for the diameter smaller than 6 μm (Figure 31a). Differences in amplitudes over sizes below 6 μm can be a result of both statistical differences in distributions averaged and/or differences in removal properties for the regions sampled. A greater concern is the much lower OPC values above about 8 μm. This is due to the fact that the upper detection limit of
the OPC did not allow for the large particles to be reconstructed even after the RH adjustment. However, this comparison provides a means to estimate the coarse particle scattering not reconstructed from the OPC data. First, the mean OPC distributions were normalized with reference to the scattering up to 8 μm. This is because despite different geographical features between Woodcock’s and my measurements would result in different scattering coefficients as an integral over diameters, the shape of sea-salt distribution should be more or less the same under a given wind speed class. The scattering losses are given as the area between the normalized OPC scattering distribution (thick blue curves) and the model scattering distribution (black curves) over 8 μm in Figure 31b, and are calculated as 16% for the 5.5 – 7.9 m/s case and 27% for the 13.9 – 17.1 m/s case. This suggests that the magnitude of the scattering underestimated from the earlier analysis may be in the order of a few tens of percent. This estimated scattering deficit could be larger or smaller than needed for a given measurement depending on the number of particles larger than 8 μm. This may depend on the different altitude, wind speed, and other meteorological processes.

9.2 NOVAM model

Part of the support and motivation for this study came from concerns over the validity of vertical scattering coefficients produced in the Navy Oceanic Vertical Aerosol Computer Model (NOVAM). NOVAM calculates scattering coefficients at 7 given altitudes up to 2000 m based on current wind speed, average wind speed over the previous 24 hours, relative humidity, and many other parameters that describe the
atmospheric conditions. Here a comparison is made between the ACE 1 and the vertical scattering coefficient calculated by NOVAM for various wind speeds. The relative humidity was set as 80%, a typical value for the ACE 1 marine air. The current wind speed parameter was set as 11 – 20 m/s for the high wind speed category, 6 – 10 m/s for the medium, and 1 to 5 m/s for the low. The average wind speed over the previous 24 hours was kept the same as the current wind speed. The resulting NOVAM vertical scattering coefficients tend to be larger than the OPC-derived scattering coefficients, especially in the medium and low wind categories (Figure 32). Comparisons may be different by stratifying the ACE 1 data into cases that best suit various other meteorological parameters used in NOVAM corresponding to the ACE 1 conditions. However, this would be a time consuming exercise beyond the scope of this thesis.

The NOVAM contains the Navy Aerosol Model (NAM) as a kernel. NAM calculates size distribution as an output. Figure 33 shows comparison of size distributions under a wind speed of 10 m/s and an RH of 80%. The ACE 1 size distribution shown in this figure is the average of all the measurement under this wind speed and RH conditions at 200 m or lower. The ACE 1 ambient size distribution is significantly smaller than the NAM size distributions under all conditions. This may be because of the frequent precipitation events observed during ACE 1 are not represented in the model, which prevent the size distribution from remaining as abundant as expected by the NAM.
10. DISCUSSION AND CONCLUSIONS

The ACE 1 and PEMT-B provided opportunities to study the marine aerosol optical properties under clean atmospheric conditions to describe their dependence on wind speed and other meteorological processes. Thermal analysis of particles at near ambient temperature (40 °C), 150 °C, and 300 °C enabled us to estimate size distributions of the volatile (mostly sulfate and organic) and refractory (mostly sea-salt) modes as well as their contributions to optical depth and cloud condensation nuclei. However, due to the poor sampling efficiency of the ACE 1 CAI inlet, determined only recently during PELTI, significant corrections and adjustments had to be made in order to use the ACE 1 data in this thesis.

As a starting point for this analysis comparisons were made for instrument performance downstream of the CAI. The OPC-derived total scattering coefficient under dry conditions was found to be about 85% of the nephelometer scattering coefficient as measured inside the aircraft. The OPC tubing loss was estimated to be small, and the corrected distribution agreed well with the particle mass measured with the internal filter sampler. The inlet loss correction resulted in an increase in the mean OPC scattering coefficient by 70% and also contributed the largest uncertainty in the resulting ambient size distribution and scattering coefficients. The OPC-derived mass, after the CAI loss correction for the ACE 1 data set, was found to be almost same as the mass measured with the external filter mass sampler for half of all the cases and smaller by a factor of 2 to 3 for the cases with highest mass loading. The large differences present at higher concentrations are expected to be due to a
significant mass contribution by the particles too large to be detected behind the CAl inlet. Comparisons with a sea-salt model also suggested that the actual sea-salt mass is larger by about a factor of two. Although those large particles dominate a significant portion of the total aerosol mass, this same sea-salt model suggested that their contribution to the scattering was a few tens of percents.

After “dry” size distributions were corrected for these sampling issues the diameter adjustment for the RH difference in the OPC and the ambient condition was applied. This resulted in an increase in the mean apparent OPC scattering coefficient by a factor of 5.2. Integration of these values over the column provided assessment of ambient optical depths due to sea-salt. Column optical depths were then estimated by estimating contributions from altitudes not reached by the aircraft. It is encouraging that after all these extensive transformations, the OPC-derived column optical depth and the sunphotometer measurements were found to be within the error bars for most of the available cases.

The average size distributions classified by the wind speed confirmed that high wind speed is associated with high sea-salt loading due to enhanced sea-salt production and vertical transport. This relationship between wind speed and sea-salt concentration holds true not only over the coarse mode but also for the accumulation mode. However, the total number of particles including both sea salt and volatile particles larger than 0.08 μm does not vary as significantly over all the wind speeds as their volume does. This implies that in these ACE 1 cases, number of the marine aerosol as potential cloud condensation nuclei (CCN) depend little on wind speed. It is still possible that the effectiveness of the marine aerosol as CCN depends on wind
speed, as the relative abundance of their sea-salt and sulfate components as well as their affinity to water do vary depending on wind speed.

This study focused on the optical properties of marine aerosol over the Southern Ocean. The average scattering coefficient increases with wind speed. During ACE 1, the median scattering coefficient showed deeper mixed layer under high wind speed, which results from increased turbulence and transport of particles from the ocean surface. Scattering was found to be almost always much smaller above 1200 m than below even in the highest wind speed case. The optical depths were calculated as integrals of the median scattering coefficients in the 200-m altitude bins. The overall aerosol optical depth, including the estimated contribution from the altitudes higher than 2000 m, was 0.063. This value is larger than 0.059, the optical depth measured on Cape Grim with the contribution from the stratosphere. This is consistent with the differences in typical wind speeds and associated scattering coefficients in the MBL between the region upwind of Cape Grim and the open ocean to the south of the island.

All these average optical properties are associated with such large variances that the wind speed alone cannot predict their instantaneous values. For example, the scattering coefficient in the lowest 200-m layer under 5 – 10 m/s can be anywhere between 10 and 150 Mm$^{-1}$. During the ACE 1 Lagrangian flight series I determined that cloud processing, wind speed history, and vertical wind speed gradient contribute to the variance. In addition, precipitating clouds appeared to scavenge most of the particles from air detrained at cloud level and helped to maintain a gradient. During periods with a weak vertical wind speed gradient and turbulence, few sea-salt
particles appear to be transported from the ocean surface to higher altitudes even under high wind speed. In addition, a natural variability of 15 Mm$^{-1}$ or 35% of the median scattering coefficient below 1000 m was observed over a horizontal distance of about 60 km. This rather large natural variability must be taken into consideration when comparing column optical properties measured during a vertical profile to average optical properties over spatial scales of 10 - 100 km observed by satellite.
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Takemura, T., H. Okamoto, Y. Maruyama, A. Numaguti, A. Higurashi, and T. Nakajima, Global three-dimensional simulation of aerosol optical thickness


Twomey, S., Atmospheric aerosols,, 1977.


Table 1. Sources of errors and their estimated magnitude.

<table>
<thead>
<tr>
<th>error source</th>
<th>magnitude</th>
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<tr>
<td>OPC calibration</td>
<td>5% for diameter and 10% for number</td>
</tr>
<tr>
<td>flow rate</td>
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<tr>
<td>sea-salt density</td>
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<tr>
<td>CAI transmission efficiency</td>
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<td>RH measurement</td>
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<td>growth factor</td>
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Table 2. Impact of each correction and adjustment expressed as the number and scattering of the mean distribution after each treatment divided by that of the raw distribution.

<table>
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<th>Correction</th>
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<th>Scattering</th>
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<td>RH Adjustment</td>
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</table>
Table 3. Number, area, volume, and scattering of the mean size distributions over the lowest 200 m during ACE 1. The data is categorized in high, medium, and low wind speed cases, which refer to wind speed greater than 10 m/s, 5-10 m/s, and lower than 5 m/s, respectively. The unit is cm$^{-3}$ for number, μm$^2$/cm$^{-3}$ for area, and μm$^3$/cm$^{-3}$ for volume. The refractory ratios (300 oC / 40 oC) are also shown.

<table>
<thead>
<tr>
<th>Wind Speed</th>
<th>&gt;0.15 μm</th>
<th>0.15 - 0.4 μm</th>
<th>0.15 - 1 μm</th>
<th>&gt;1 μm</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Number</td>
<td>Area</td>
<td>Volume</td>
<td>Scattering</td>
</tr>
<tr>
<td>all</td>
<td>high</td>
<td>medium</td>
<td>low</td>
<td>all</td>
</tr>
<tr>
<td>40 oC</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>150 oC</td>
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<td></td>
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</tr>
<tr>
<td>300 oC</td>
<td></td>
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<tr>
<td>300 oC / 40 oC</td>
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</tr>
</tbody>
</table>

...
Table 4. Number, area, volume, and scattering of the mean size distributions over the lowest 200 m during PEMT-B. The data is categorized in high, medium, and low wind speed cases, which refer to wind speed greater than 10 m/s, 5-10 m/s, and lower than 5 m/s, respectively. The unit is cm$^{-3}$ for number, μm$^2$/cm$^{-3}$ for area, and μm$^3$/cm$^{-3}$ for volume. The refractory ratios (300 oC / 40 oC) are also shown.

<table>
<thead>
<tr>
<th>&gt;0.15 um</th>
<th>Number</th>
<th>Area</th>
<th>Volume</th>
<th>Scattering</th>
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<tr>
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<td>0.11</td>
<td>0.12</td>
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</tr>
</tbody>
</table>

| 0.15 - 0.4 μm | Number | Area | Volume | Scattering |
| Wind Speed | all | high | medium | low | all | high | medium | low | all | high | medium | low |
| 40 oC | 138.9 | 148.9 | 127.6 | 98.6 | 28.9 | 28.6 | 28.2 | 23.4 | 1.4 | 1.4 | 1.4 | 1.2 | 4.6 | 4.6 | 4.8 | 4.5 |
| 150 oC | 125.0 | 128.3 | 121.8 | 100.1 | 24.4 | 23.2 | 25.8 | 23.3 | 1.1 | 1.0 | 1.2 | 1.2 | 3.6 | 3.2 | 4.2 | 4.2 |
| 300 oC | 10.8 | 9.4 | 11.7 | 13.1 | 2.3 | 2.0 | 2.6 | 2.8 | 0.1 | 0.1 | 0.1 | 0.1 | 0.4 | 0.3 | 0.4 | 0.3 |
| 300 oC / 40 oC | 0.06 | 0.06 | 0.09 | 0.13 | 0.08 | 0.07 | 0.09 | 0.12 | 0.08 | 0.07 | 0.09 | 0.11 | 0.06 | 0.07 | 0.09 | 0.08 |

| 0.15 - 1 μm | Number | Area | Volume | Scattering |
| Wind Speed | all | high | medium | low | all | high | medium | low | all | high | medium | low |
| 40 oC | 164.0 | 163.9 | 164.0 | 143.5 | 51.6 | 43.7 | 59.9 | 68.2 | 3.6 | 2.8 | 4.4 | 5.7 | 17.5 | 13.9 | 22.9 | 13.1 |
| 150 oC | 147.1 | 141.8 | 153.2 | 142.2 | 44.7 | 36.3 | 53.3 | 64.6 | 3.1 | 2.4 | 3.9 | 5.3 | 15.2 | 11.9 | 20.1 | 11.4 |
| 300 oC | 14.8 | 13.3 | 16.2 | 17.4 | 7.7 | 7.4 | 8.0 | 7.8 | 0.8 | 0.8 | 0.8 | 0.7 | 4.7 | 4.9 | 4.6 | 3.2 |
| 300 oC / 40 oC | 0.09 | 0.08 | 0.10 | 0.12 | 0.15 | 0.17 | 0.13 | 0.11 | 0.21 | 0.27 | 0.17 | 0.12 | 0.27 | 0.35 | 0.20 | 0.24 |

| > 1 μm | Number | Area | Volume | Scattering |
| Wind Speed | all | high | medium | low | all | high | medium | low | all | high | medium | low |
| 40 oC | 5.1 | 5.9 | 4.4 | 3.3 | 68.8 | 79.8 | 56.5 | 47.2 | 33.4 | 38.9 | 26.8 | 25.3 | 40.7 | 47.3 | 33.8 | 13.5 |
| 150 oC | 4.8 | 5.6 | 3.9 | 3.2 | 63.2 | 74.6 | 50.1 | 42.8 | 30.1 | 35.8 | 23.4 | 21.5 | 37.4 | 44.3 | 30.0 | 12.4 |
| 300 oC | 4.5 | 5.3 | 3.7 | 2.6 | 59.4 | 71.6 | 47.4 | 37.0 | 28.6 | 34.8 | 22.3 | 19.2 | 35.2 | 42.5 | 28.3 | 10.9 |
| 300 oC / 40 oC | 0.87 | 0.90 | 0.85 | 0.79 | 0.86 | 0.90 | 0.84 | 0.78 | 0.86 | 0.90 | 0.83 | 0.76 | 0.86 | 0.90 | 0.84 | 0.81 |
Figure 1. Example of particle number, area, and volume distributions.
Figure 2. (a) Growth factor for the marine sea-salt aerosol as a function of relative humidity after Tang et al. [1997]. (b) f(RH) as functions of relative humidity after Clarke et al. [personal communication]. (c) Refractive index as a function of relative humidity after Tang et al. [1997].
Figure 3. Scattering distribution (black) for particles with a refractive index of 1.57 at 550 nm. It is given as a product of the cross sectional area (magenta) of the particles and scattering efficiency (thin blue). For the typical size distribution such as shown here particles of 0.1 – 10 μm are most efficient on a mass basis in scattering as indicated by the thick blue curve.
Figure 4. Vertical nephelometer dry scattering profile at three wavelengths (450, 550, and 700 nm) and the Angstrom exponent inferred from the 450 and 700 nm measurement during ACE 1 flight 26, 00:15 – 00:28 over the Southern Ocean.
Figure 5. ACE 1 flight tracks at 2000 m or lower. Also shown are the flight numbers. Black ‘x’s indicate the R/V Discoverer positions during the sunphotometer measurements.
Figure 6. PEMT-B flight tracks at 2000 m or lower.
Figure 7.  (a) Passing efficiency of the internal sampler. The overall efficiency (black dots) is given as a product of the efficiencies due to aircraft plumbing (triangles) and impactor (crosses). The efficiency curve against geometric diameter (red dots) was applied in the actual correction of the measured mass. (b) Transmission efficiency for the nephelometer impactor and the OPC tubing.
Figure 8. Scattering efficiency seen by nephelometer (red) for a refractive index of 1.58 at 550 nm as compared with the normal scattering efficiency (blue).
Figure 9. Airborne scattering closure after tubing loss correction at 450, 550, and 700 nm. Circles are for uncorrected OPC scattering, and squares are those corrected for the OPC tubing loss.
Figure 10. Comparison of Angstrom exponent inferred from the OPC size distribution and the nephelometer scattering coefficient.
Figure 11. Mass closure inside (green circles) and outside (blue dots) the aircraft. For the internal comparison, the OPC-derived dry sea-salt mass before (black) and after (green) the tubing loss correction is compared to the dry sea-salt mass measured by the internal sampler (corrected for the transmission efficiency, Figure 7a). The R² values for both green and black dots are 0.85. For the external comparison, the OPC-derived dry sea-salt mass after the tubing and CAI loss corrections is compared to the dry sea-salt mass measured by the external sampler. The error bars are larger for the external OPC mass than for the internal OPC mass, as the CAI transmission efficiency is the largest source of uncertainty (Figure 12).
Figure 12. CAI transmission efficiency for ACE 1 and Solid Diffuser transmission efficiency for PEMT-B, both plotted against aerodynamic diameters. The large diameter end of the CAI transmission efficiency curve was modified as explained in the text. The area with the diagonal lines associated with each curve indicates the variability in the transmission efficiency curves obtained during PELTI.
Figure 13. Impact of each of the corrections and the adjustment, shown as average number and scattering distributions of the marine aerosol during ACE 1 (a) and PEMT-B (b).
Figure 14. Closure of the reconstructed scattering coefficients inferred from the OPC and the nephelometer measurements. Both of these data are the results of the transformations as explained in the text.
Figure 15. Example of column closure between the OPC-derived and the nephelometer-derived ambient scattering coefficient, encountered during ACE 1 flight 17, 02:58 - 03:11 (UTC), November 28, 1995.
Figure 16. Average number and volume distributions below 200 m for all, high, medium, and low wind speed categories during ACE 1 and PEMT-B. For ACE 1, the RDMA unheated distribution is also shown.
Figure 17. Average number and volume distributions of refractory particles during ACE 1 (expanded from Figure 16).
Figure 18. Average scattering distributions below 200 m for all, high, medium, and low wind speed categories during ACE 1 and PEMT-B.
Figure 19. Average number and volume distributions over 200 – 600 m for all, high, medium, and low wind speed categories during ACE 1 and PEMT-B. For ACE 1, the RDMA unheated distribution is also shown. Note that the y scales are different from Figure 16 and 20.
Figure 20. Average number and volume distributions over 600 – 1000 m for all, high, medium, and low wind speed categories during ACE 1 and PEMT-B. For ACE 1, the RDMA unheated distribution is also shown. Note that the y scales are different from Figure 16 and 19.
Figure 21. Vertical ambient scattering profile inferred from the OPC unheated size distribution during ACE 1.
Figure 22. Vertical ambient scattering profile due only to sea-salt particles inferred from the OPC 300°C size distribution during ACE 1.
Figure 23. Vertical ambient scattering profile inferred from the nephelometer measurements during ACE 1.
Figure 24. Optical depth closure. The sunphotometer measurement was done aboard R/V Discoverer while the aircraft flew nearby. The error bars for the OPC-derived optical depth is calculated based on the estimated error sources listed in Table 1. Also shown is the optical depth measured at Cape Grim and averaged over December 1986 – 1999 except 1991. The contribution from stratospheric aerosol has been added.
Figure 25. Median optical depth in each wind speed classes. Stratospheric contribution (light blue) has been added to the OPC-derived differential optical depth integrated over 0 - 2000 m (thick blue).
Figure 26. ACE 1 scattering coefficient (left axis) and wind speed (right axis) averaged over 1° latitude. Tasmania is located between 40 – 44°S.
Figure 27. Flight path and wind speed during ACE 1 Lagrangian A series.
Figure 28. Change in vertical size distribution profile during the Lagrangian A series during ACE 1. The blue, red, and black curves represent the OPC size distributions at 40, 150, and 300 °C, respectively.
Figure 29. Vertical scattering and wind speed profiles during the Lagrangian B series in ACE 1.
Figure 30. a) Example of the sets of a descending profile and several horizontal legs, used for the natural variability analysis. b) Frequency of the difference in scattering coefficient measure during the initial descending profile and the horizontal legs.
Figure 31. Modeled and measured size distributions under two different wind speed categories over 500 - 800 m.
Figure 32. Modeled and measured vertical scattering profiles. The green dots represent the median scattering coefficient during ACE 1.
Figure 33. Modeled and measured size distributions. NAM results and the OPC-derived ambient size distributions are shown in number (a), area (b), volume (c), and scattering (d). Three different modes in NAM are also shown with thin dotted curves.